Proceedings of the National Seminar & Exhibition on Non-Destructive Evaluation
NDE 2009, December 10-12, 2009

Microstructural Characterization of Precipitation Process in a Nickel-Alloy by Non-linear Ultrasonic

Avijit Metya, G.V.S.Murthy, H.N. Bar and N.Parida
MST Division, National Metallurgical Laboratory (CSIR)-Jamshedpur-831 007, India

Abstract
The nonlinear ultrasonic technique, using the amplitude ratio of higher harmonic frequencies and fundamental frequency, has been found to be strongly sensitive to the microstructure of bulk materials. It was reported earlier that in Al 2024 alloy the nonlinearity parameter increased with the generation of coherency precipitates. Similarly, Hurley et al reported that the nonlinear parameter linearly increases as a function of inhomogeneous strain due to the generation of a precipitate in low alloy steel. In contrast to these studies, in which researchers have studied ultrasonic nonlinearity in the context of single crystals and simple metals, we would like to study structural materials for the purpose of structural health monitoring. In order to characterize the material properties in facilities and during operation, one needs to understand the relation between the material degradation of structural materials and the features of the NDE parameters. Therefore, in the present study we attempted to assess the thermal degradation in one such structural material namely: Nimonic-263, nickel based precipitation hardenable alloy using the nonlinear technique. From the present study it was found that the response of the non-linear ultrasonic parameter b is faster and larger compared to the normal velocity measurements.

Introduction

During the lifetime of a plant component, the structural material is exposed to various types of loading and temperature. Such unforeseen conditions may have positive or, more worryingly, negative effects on the structural properties of the material. Typical variations in the material properties cannot be detected in situ using conventional destructive methods. Therefore, the development of nondestructive evaluation (NDE) techniques is of utmost importance for detecting defects and evaluating material properties in a structural health monitoring (SHM) program. Towards this aim, we note that the ultrasonic nonlinear technique, using the amplitude ratio of fundamental frequency and higher harmonic frequencies, has been found to be strongly sensitive to the microstructure of bulk materials [1–3]. The nonlinear elasticity of a solid is related to the forces acting between the atoms in a crystal lattice. These interatomic potentials (e.g., the Lennard-Jones potential) can be characterized by the well depth and equilibrium interatomic separation. Interatomic potentials in real crystals are not harmonic, and the anharmonicity can be treated through higher order elastic theory by adopting a continuum approximation. The potentials are determined by measurement of the energy of the crystal and of its lattice spacing. When an ultrasonic sinusoidal wave of a given frequency and of sufficient amplitude is introduced into an anharmonic solid, the fundamental wave may be distorted as it propagates so that the second and higher harmonics of the fundamental frequency will be generated. Measurement of the amplitude of the second harmonic as a function of the amplitude of the fundamental frequency has been carried out by Breazeal and Tompson [4] for the purpose of understanding the material anharmonicity. Breazeale and Ford [5] studied the nonlinear behavior of ultrasonic waves in single crystals of copper, which had been neutron irradiated, via finite amplitude distortion in order to observe the nonlinear properties of the crystal lattice alone. Hikata et al. [6] investigated the dislocation contribution to the nonlinearity using a single crystal of aluminum, and they reported that the amplitude of the second harmonic increased as a function of the tensile stresses. Cantrell and Yost [7] presented a model in which the dependence of acoustic harmonic generation in polycrystalline solids on the coherency strains results from the lattice mismatch at the interface between the matrix and the second phase in Al 2024 alloy. They reported that the nonlinearity parameter increased with the generation of coherency precipitates. Hurley et al. [8] reported that the nonlinear parameter linearly increases as a function of inhomogeneous strain due to the generation of a precipitate in low alloy steel. In contrast to these studies, in which researchers have studied ultrasonic nonlinearity in the context of single crystals and simple metals, we would like to study structural materials for the purpose of SHM. In order to characterize the material properties in facilities and during operation, one needs to understand the relation between the material degradation of structural materials and the features of the NDE parameters. Therefore, in the present study we attempted to assess the thermal degradation in Nimonic-263, nickel based precipition hardenable alloy using the nonlinear technique.

The nickel-base superalloy Nimonic 263 was developed and introduced by Rolls Royce in 1960. The alloy can be age-hardened by the controlled precipitation of intragranular sub-microscopic γ' phase, which contributes to creep resistance by acting as a barrier to dislocation movement. The presence of carbon leads to the formation of a series of carbide phases: (a) Intragranularly occurring primary carbides, nitrides or carbonitrides of the general form M(C, N), where M is usually titanium, and (b) chromium-rich grain boundary carbides like

M_7C_3 and $M_{23}C_6$ [9]. It is designed primarily for use in the stationary components like combustion chamber, casing, liner, exhaust ducting, bearing housing and many others. These components are fabricated from plate/sheet of this alloy. The formability and weldability of the alloy hence become an important factor for the fabrication. The standard heat treatment for Nimonic 263 is solution annealing at 1423 K for 2 h followed by ageing at 1273 K for 8 h [9]. In the optimum heat treated condition, the microstructure of the alloy shows a fine discontinuous precipitation of carbides at the grain boundaries and precipitation of γ' intermetallic in the matrix [9]. The shape and size of γ' precipitates cannot be resolved in an optical microscope. However, electron microscopic studies show that the mean particle diameter is about a few nanometers. The γ' phase that forms is metastable in nature because of the high titanium to aluminum ratio, the stable precipitate being η-Ni_3Ti. On prolonged exposure at temperatures in excess of 1073 K, the γ' phase gradually coarsens and acicular η begins to form [9]. The primary objective of the solution treatment is to dissolve the precipitated phases, mainly γ' and carbides, prior to their controlled precipitation during ageing. Ageing treatments are primarily concerned with the precipitation of the hardening phase, normally γ' in a form to obtain the required mechanical properties. The response to the ageing treatment depends on time and temperature used for the heat treatment.

Experimental

Table 1 shows the chemical composition of the alloy used in the present study. A set of Nimonic 263 specimens of dimensions 20 mm×20 mm×10 mm was solution annealed (SA) at 1423 K for 1 h followed by water quenching. The SA specimens were thermally aged for 1, 2, 4, 6 and 8 h at 923 K and 1, 2, 4, 6, 25, 50 and 75 h at 1073 K, followed by water quenching. The samples are designated as ‘S’ for solutionized only, ‘A1’ solutionized and aged at 923 K for 1 h up to ‘A5’ solutionized and aged at 923 K for 8 h. Similarly ‘B1’ represents solutionized and aged at 1073 K for 1 h up to ‘B7’ solutionized and aged at 1073 K for 75 h.

The ultrasonic nonlinearity measurement system primarily consisted of a high power pulse generator (RAM10000, RITEC Inc.), a high power attenuator(RA-31), a high power 50Ω termination and a high powered Battenuator. A longitudinal piezoelectric transducer, with a nominal frequency of 5MHz, was used to generate the fundamental wave. A broad band 10MHz transducer was used to measure the second higher harmonic wave. This detected wave was digitally processed using an amplitude spectrum Fast Fourier Transformation to obtain the amplitudes, A_1 and A_2 at the fundamental frequency and second harmonic frequency, respectively. If the attenuation is not negligible, β may be influenced by the material attenuation (i.e.,liquid, gas,and tissue etc.). In this study, we have driven the nonlinear wave equation considering that the attenuation is negligible. Also, the difference in attenuation coefficient at the fundamental frequency and second harmonic frequency was not large; moreover, heat treated specimens showed no change in attenuation coefficient at the frequency range (5–10 MHz). Therefore, the correction of attenuation was not carried out.

Vicker’s hardness measurements were carried out using an AFFRI VRSD 270 hardness tester at a load of 30 kg. An average of five readings is reported here. A maximum scatter of ±5 VHN was obtained in hardness measurements in any specimen. Scanning electron microscopy microscopy was carried out to study the precipitation behavior to corroborate the ultrasonic velocity and hardness measurements.

3. Results and discussion

Table 2 shows the variations in hardness with ageing time at 923 and. The solution annealed specimen exhibited the lowest hardness (182 HV) and it increased with ageing at 923 K and 1073 K beyond 1 h of ageing. Upon ageing at 923 K, hardness was found to increase continuously up to the maximum duration of ageing (8 h) used in the present investigation. Upon ageing at 1073 K, the hardness increased rapidly up to 6 h (277 HV), followed by a subtle increase up to 50 h (297 HV) and then by a small decrease at 75 h (277 HV). The increase in the hardness at both temperatures is attributed to the precipitation of coherent γ' intermetallic phase. The shorter incubation period and faster increase in hardness during the initial period of ageing (up to 6 h) at 1073 K as compared to 923 K is attributed to the faster kinetics of precipitation at higher temperature (1073 K). The decrease in hardness upon ageing at 1073 K for 75 h is attributed to the coarsening of fine coherent γ' phase and its conversion to acicular η phase. The results obtained are in line with the time temperature transformation (TTT) diagram reported in literature for the precipitation of γ' intermetallic phase in this alloy. The variation in hardness with ageing time and temperature is also confirmed by the scanning electron microscopy studies. Fig. 1 (a and b) show the microstructures corresponding to the specimens thermally aged 1073 K for 6 h and 1073 for 50 h, respectively. The micrograph (Fig. 4A) of the specimen aged at 1073Kfor 6 h clearly shows the precipitation of the γ' phase. The micrograph (Fig. 1b) of the specimen thermally aged at 1073 K for 50h exhibits extensive precipitation of γ' intermetallic phase alongwith the grain boundary carbides.

There are several theories of precipitation hardening. According to the coherent lattice theory [10], the alloy remains in a supersaturated condition after solution treatment and quenching, with the solute atoms distributed at random in the lattice structure. The process of precipitation of intermetallic phases upon ageing the solution annealed alloy can be divided into two stages: (I) an incubation period, when the excess solute atoms tend to migrate to certain

Table 1 : Chemical composition of Nimonic-263

Element	*C*	*Co*	*Cr*	*Mo*	*Ti*	*Al*	*Cu*	*O*	*N*	*Ni*
Wt%	0.06	19.6	20.1	6.0	2.0	0.4	0.001	0.003	0.006	Bal

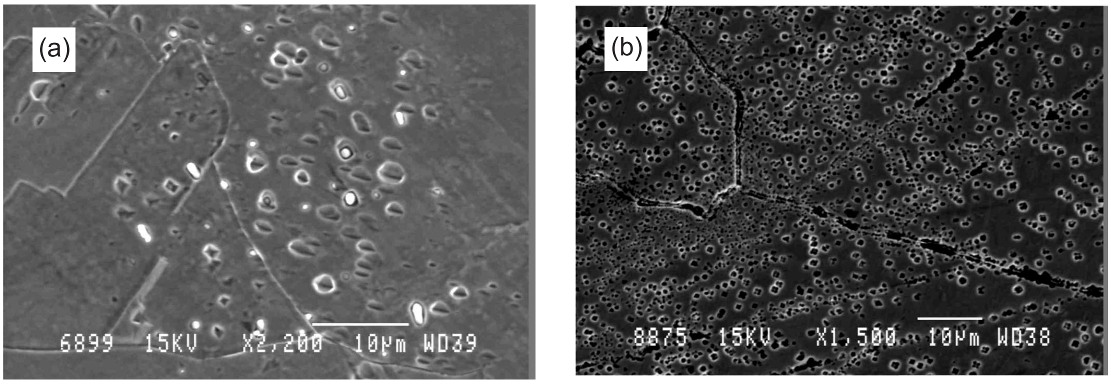

Fig. 1 : Scanning electron micrographs of Ni-263 specimens solution annealed at 1423 K for 1 h followed by ageing at (a) 1073 K for 6 h showing precipitation of γ' and (b) 1073 K for 50 h showing extensive precipitation of globular γ'.

Table 2 : β-parameter and Hardness data of the sample Nimonic-263

Sample	β *(NL-parameter)*	*Hardness (Hv)*
Solutionised	0.075966	182
Aged 923K/1 h	0.39531	183
Aged 923K/2 h	1.4761	193
Aged 923K/4 h	0.093795	209
Aged 923K/6 h	0.00668535	215
Aged 923K/8 h	0.027948	236
Aged 1073K/1 h	0.007651	224
Aged 1073K/2 h	0.0209085	241
Aged 1073K/4 h	0.00742	266
Aged 1073K/6 h	0.0062637	277
Aged 1073K/25 h	0.062590667	285
Aged 1073K/50 h	0.006167667	297
Aged 1073K/75 h	0.011356267	277

crystallographic planes, forming clusters or embryos of the precipitate and (II) precipitation, when these clusters form an intermediate crystal structure or transitional lattice, maintaining registry (coherency) with the lattice structure of the matrix. This phase will have lattice parameters different from those of the solvent, and as a result of the atom matching (coherency) there will be considerable distortion of the matrix.

This distortion (strain free) extends over a distance more than the size of a discrete (precipitate) particle. It is this distortion that interferes with the movement of dislocations and accounts for the increase in hardness and strength during ageing. The first stage of the precipitation (i.e., incubation) does not influence the hardness; however, it drastically influences the elastic moduli of the material and in-turn the ultrasonic velocities. The depletion of the precipitate-forming elements from the matrix leads to an increase in the modulus of the matrix [10].

References

1. Cantrell J H, Yost WT. Determination of precipitate nucleation and growth rates from ultrasonic harmonic generation, *Appl Phys Lett.*, **77**(13) (2000) 1952–4.
2. Kim J Y, Jacobs L J and Qu J, Experimental characterization of fatigue damage in a nickel-base superalloy using nonlinear ultrasonic waves, *J. Acoust. Soc. Am.*, **120**(3) (2006) 1266–73.
3. Kim C S, Kim Y H and Kim I H. Ultrasonic linear and nonlinear parameters in cyclically deformed Cu and Cu-35Zn alloy, *Key Eng Mater.*, (2005) 297–300 (3): 2134–9. (5):77–8
4. Breazeale M A and Thompson D O. Finite-amplitude ultrasonic wave in aluminum. *Appl. Phys. Lett.*, **3**(5) (1963) 77–8.
5. Breazeale M A and Ford J, Ultrasonic studies of the nonlinear behavior of solids. *J. Appl. Phys.*, **36**(11) (1965) 3486–90.
6. Hikata A, Chick B B and Elbaum C, Effect of dislocations on finite amplitude ultrasonic waves in aluminum. *Appl. Phys. Lett.*, **3**(11) (1963) 195–7.
7. Cantrell J H and Yost W T, Effect of precipitate coherency strains on acoustic harmonic generation. *J. Appl. Phys.*, **81**(7) (1997) 2957–62.
8. Hurley D C, Balzar D and Purtscher P T, Nonlinear ultrasonic assessment of precipitation hardening in ASTM A710 steel, *J. Mater. Res.*, **15**(9) (2000) 2036–42.
9. Singh P N and Singh Vakil, Influence of ageing treatment on work-hardening behaviour of a Ni-base superalloy, *Scr. Mater.*, **34** (1996) 1861–5.
10. Avener S H, Introduction to physical metallurgy. Second Ed. Tokyo: Mcgraw Hill; (1974) p. 192.